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AMORPHOUS MAGNETISM IN F. C. C. VICALLOY II

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INTRODUCTION

Although the occurrence of superparamagnetism has been inferred in many binary alloy systems in which one of the alloy components is non-magnetic (Beck, 1971), relatively few investigations of this phenomenon have been carried out on concentrated ternary alloys. In this report we present the results of magnetization and Mossbauer experiments which give evidence for superparamagnetism in the quenched face-centered-cubic (γ) phase of the ternary system $Fe_{0.34}Co_{0.52}V_{0.14}$ (Vicalloy II). While the magnetization data agree with the limited data of Nesbitt et al (1967), our more complete study shows clearly that this alloy does not acquire long-range ferromagnetic order, nor does it indicate either antiferromagnetic long-range ordering or mictomagnetic behavior, down to 4.2° K. Our data can best be described as resulting from ferromagnetic clusters in a weakly magnetic lattice.

The Mossbauer data show a slightly broadened single line spectrum of 300° K which broadens with decreasing temperature without development of resolvable hyperfine splitting down to 4.2° K. We attribute the broadening to relaxation effects associated with magnetic cluster coalescence.

RESULTS AND ANALYSIS

Splat cooled samples of Vicalloy II were obtained from R. Willens of the Bell Telephone Laboratory, Murray Hill, N. J. Magnetometer measurements were performed with a vibrating sample magnetometer mounted in a large iron electromagnet. Calibration was with an iron sample and the effect of sample images in the pole pieces was included in data reduction. The data obtained are shown in figure 1.

These data are analyzed using the modified Langevin equation:

$$\sigma = \chi \cdot H + N\bar{\mu}\zeta\left(\frac{\bar{\mu}\cdot H}{kT}\right)$$

where σ is sample moment per gram (ergs/gm·Oe), χ is the high field susceptibility (ergs/cm·O_e^2), N is cluster number per gram, $\bar{\mu}$ is average magnetic moment per cluster (ergs/Oe) and $\zeta(\bar{\mu}H/kT)$ is the Langevin function. The results of this analysis are shown in figure 2 as a plot of $\bar{\mu}$ and N versus temperature. The high field susceptibility was temperature independent with the value $\chi = 0.35 \pm 0.1\text{x}10^{-3}$ ergs/gm·Oe_e^2.

An estimate of the magnetic moment per magnetic ion can be made using the parameters of equation (1). The result is shown in figure 3 where the average moment in Bohr magnetons per magnetic ion ($\overline{\mu}_{at}$) is plotted against temperature.

Mossbauer data were taken with a constant acceleration transmission mode experiment. Absolute velocity calibration was obtained by multiplexing nuclear data with a laser-interferometer fringe counter. The source was Co^{57} in palladium. With the exception of room temperature data, all data were taken through a cryostat with 1 mm of beryllium in the radiation beam. Parasitic nuclear absorption effects in the cryostat were corrected. Detection was by CO_2Kr proportional counter. No parasitic absorption was observed in the beryllium window of the detector. Sample temperature was controlled within 0.5° K or better for all data below room temperature.

Mossbauer data at room temperature and 4.2° K are shown in figures 4 and 5. Error bars indicate range of ± 1 standard deviation in count away from resonance. Data are analyzed by least squares with a single Lorentzian line. The parameters of fit are shown in the respective figures. Data at 4.2° K are not fit well by a single Lorentzian.

The large increase in line broadening at 4.2° K in the absence of resolvable hyperfine splitting suggests the existence of a change in the short range magnetic order. To determine the ordering temperature a thermal analysis was performed. The transmitted flux of the 14 keV line was measured at zero Doppler velocity in the temperature range of 270° K to 4.2° K. In order to obtain reproducible measurements at each temperature the observed transmitted flux at 14 keV was normalized by the observed transmitted flux at 21 keV. The results are shown in figure 6.

To identify more clearly the broadening indicated by figure 6 additional spectra were taken at 150°, 75°, and 25° K. The results are shown in figure 7. The single line Lorentzian parameters obtained by least squares fit to the data are tabulated in Table 1.

DISCUSSION

The data presented above can be described in terms of a superparamagnetic cluster model. In this model local composition variations produce local regions, which at sufficiently low temperatures, become magnetically ordered. Depending upon local composition, these regions, called clusters, possess a unique magnetic ordering temperature and a unique saturation magnetization at 0° K. An exchange coupling is allowed between clusters in close proximity such that individual clusters may coalesce to form larger clusters. The largest clusters are assumed to be small compared to the size of magnetic domains.

Consider the cluster analysis data of figure 2. At 300° K it is assumed that some magnetically ordered clusters exist and are well

separated by the remaining paramagnetic material. In the temperature range of 300° to 200° K additional clusters with ordering temperatures in this range contribute to the magnetization and increase the number of clusters per gram. Since these clusters have smaller moments than clusters which order above 300° K, they decrease the average moment per cluster. This process of magnetic cluster formation is thought to exist over the entire range of the measurements.

In the temperature region of 200° to 50° K some clusters which exist are able to coalesce to form larger ordered regions. Sufficient numbers of clusters are lost by this process to more than compensate for the number of new clusters formed in this temperature range. Consequently, the number of clusters per gram decreases and the average moment per cluster increases.

Clusters which become ordered at low temperatures are expected to have the smallest values of magnetic moment. Consequently, the exchange interaction between these clusters and the coalesced clusters may become sufficiently weak at some low temperature to favor the existence of the individual small cluster over continued growth of the coalesced cluster. The data suggest this may occur below 50° K where an increase in cluster number and a decrease in cluster moment begins.

Mossbauer thermal analysis data (figure 6) and spectra (figures 4, 5, and 7) suggest the onset of a change in the short range magnetic ordering near 150° K. This transition temperature is slightly below the transitions shown in figure 2 (i.e., 175° and 200° K) for the onset of cluster coalescence.

The broadening observed in the Mossbauer data is a consequence of a distribution of hyperfine fields at the iron nucleus. In terms of the model given above the paramagnetic line at 300° K is slightly broadened due to the existance of clusters with relaxation times somewhat less than the nuclear lifetime. The Mossbauer line width does not change appreciably from 300° to 150° K since the clusters ordered in this temperature region have smaller moments (and relaxation times) than clusters ordered above 300° K.

Clusters which coalesce at temperatures between 150° to 200° K do not produce clusters with relaxation times long enough for detection by the lifetime of the iron nucleus. Since the coalescence process will produce an abrupt change in the relaxation time of the final cluster, this suggests that coalescence in this temperature range is not between clusters having the larger values of magnetic moment and relaxation times.

In the temperature range 150° to 50° K the coalescence of clusters produces a significant number of clusters with relaxation times comparable to the nuclear lifetime. At temperatures below 50° K these clusters

produce larger hyperfine fields at the iron nucleus due to the temperature dependence of cluster spin relaxation. Clusters with ordering temperatures in the range 0° to 50° K have relaxation times too short to contribute to the nuclear hyperfine field.

CONCLUSIONS

The magnetometer and Mossbauer data clearly show that splat-cooled F.C.C. Vicalloy II does not exhibit long-range magnetic order. The short-range magnetic ordering is shown to be temperature dependent with clear indication of coalescence of magnetic clusters over a limited temperature range. The model presented reasonably accounts for the observed effects and indicates this ternary system is superparamagnetic down to 4.2° K. This finding is quite unusual for an alloy with 86 atomic percent magnetic ions. The reason for this unusual situation is found in the electronic structure of the individual cluster which will be the subject of a subsequent report.

REFERENCES

1. Beck, Paul A.: Some Recent Results on Magnetism in Alloys. Metallurgical Trans., vol. 2, no. 8, Aug. 1971, pp. 2015-2024.

2. Nesbitt, E. A.; Willens, R. H.; Williams, H. J., and Sherwood, R. C.: Magnetic Properties of Splat-Cooled Fe-Co-V Alloys. Jour. Appl. Phys., vol. 38, no. 3, Mar. 1967, pp. 1003-1004.

Temperature, °K	Line Width (FWHM), mm/sec	Isomer Shift, mm/sec
300	0.410	-0.273
150	0.463	-0.193
75	0.741	-0.153
25	1.390	-0.114
4.2	1.852	-0.091

Table 1: Parameters of least squares fit to Mossbauer spectra shown in figures 4, 5, and 7. Isomer shift is relative to Co^{57} in palladium. Line width is full width at half-maximum amplitude.

MAGNETIC MOMENT PER GRAM vs. APPLIED FIELD

Splat Cooled Vicalloy II

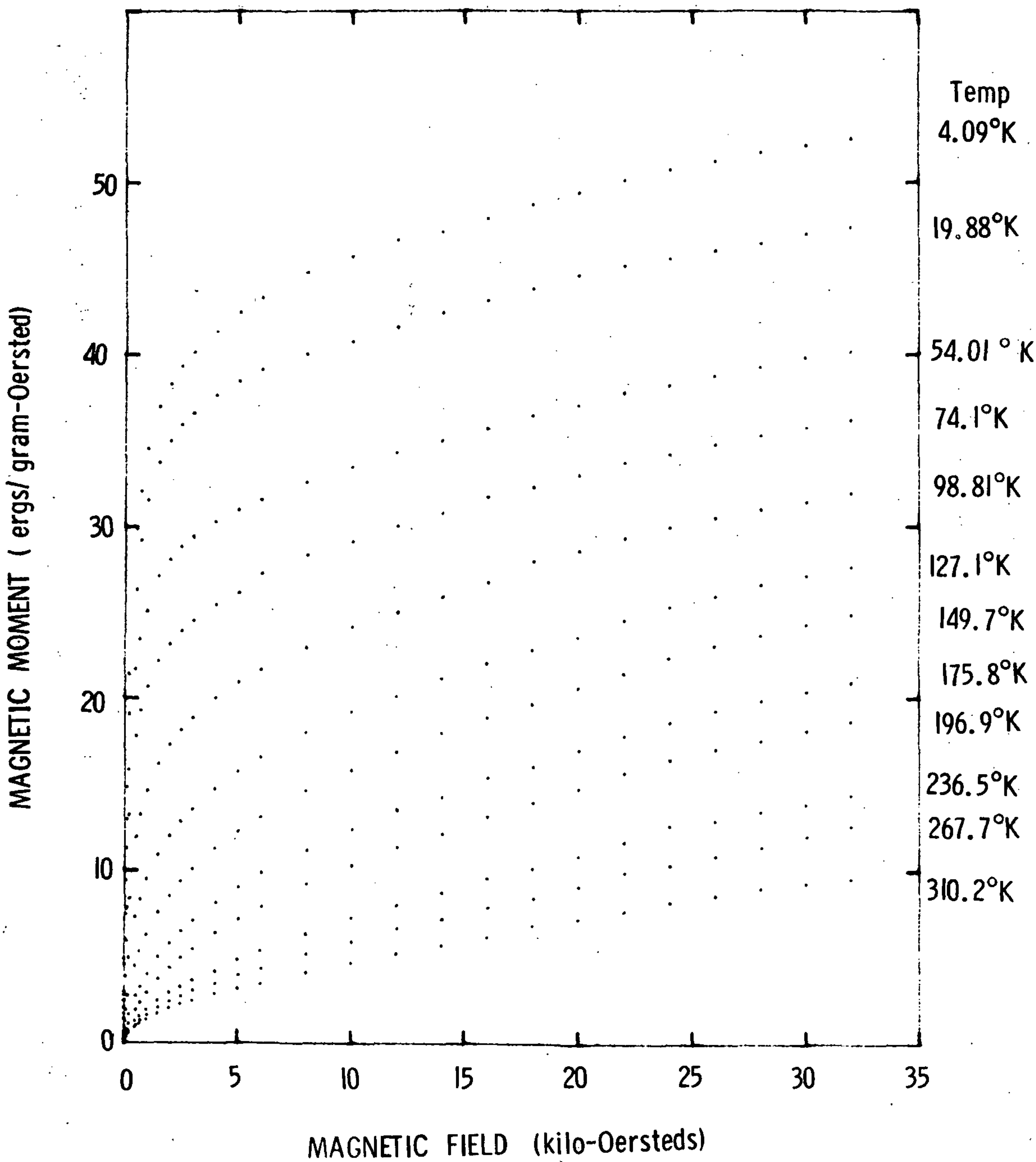


Figure I

LEAST SQUARES FIT PARAMETERS FOR MODIFIED LANGEVIN EQUATION vs. TEMPERATURE

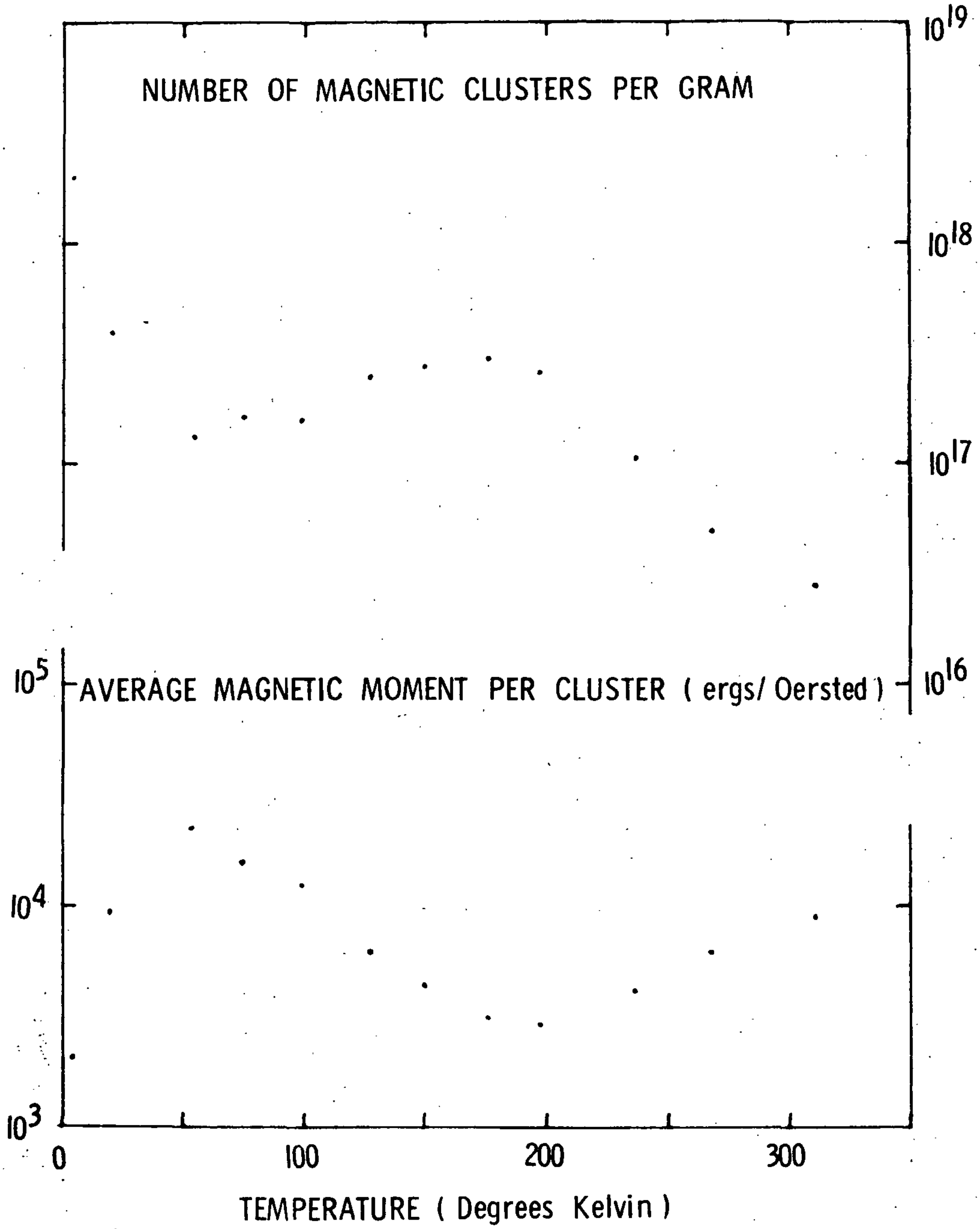


Figure 2

AVERAGE MAGNETIC MOMENT PER MAGNETIC ATOM
vs. TEMPERATURE
Splat - Cooled Vicalloy II

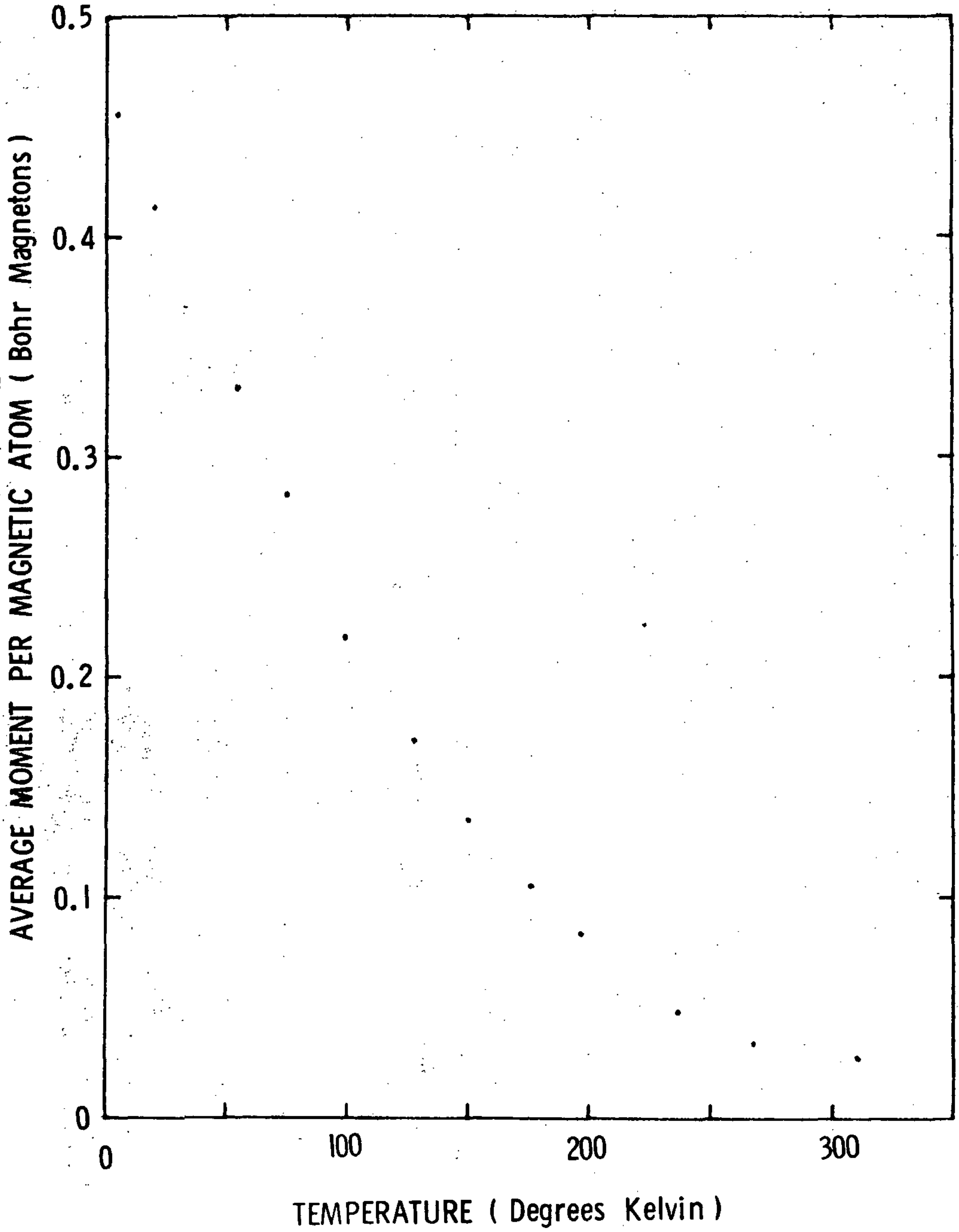


Figure 3

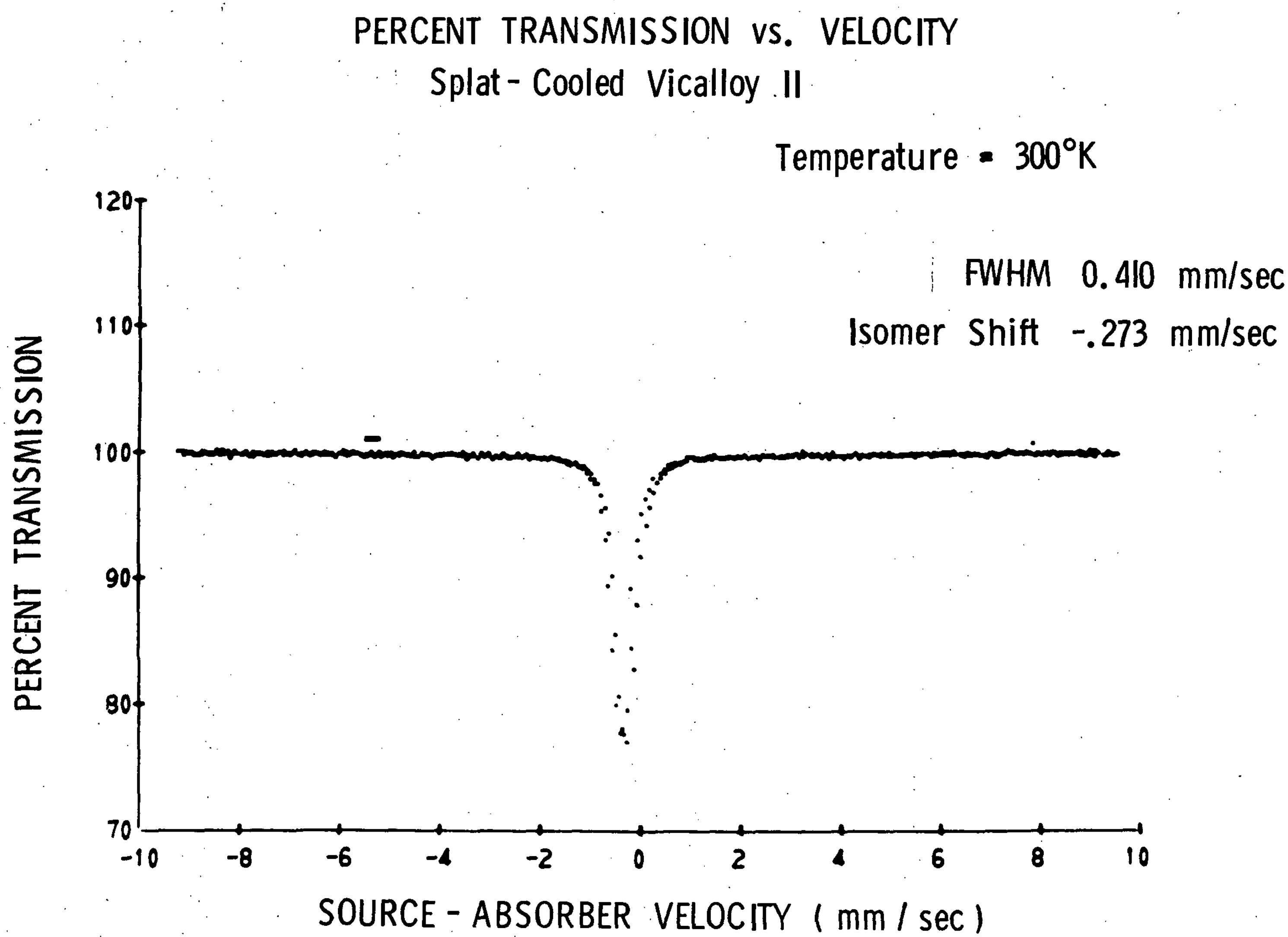
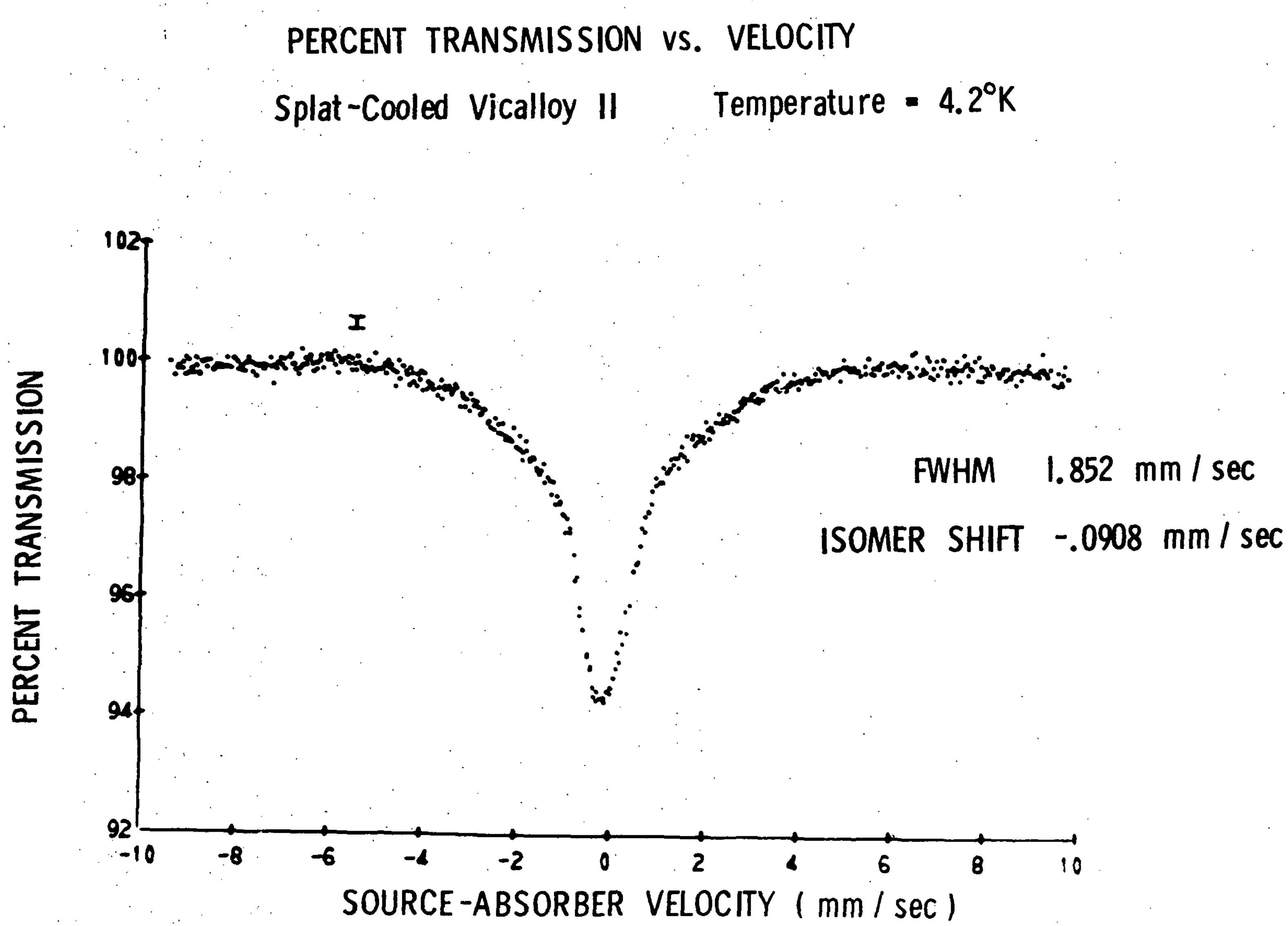

PERCENT TRANSMISSION vs. VELOCITY
Splat - Cooled Vicalloy II
Temperature = 300°K
FWHM 0.410 mm/sec
Isomer Shift -.273 mm/sec
PERCENT TRANSMISSION
120
110
100
90
80
70
-10
-8
-6
-4
-2
0
2
4
6
8
10
SOURCE - ABSORBER VELOCITY (mm / sec)

Figure 4



PERCENT TRANSMISSION vs. VELOCITY
Splat-Cooled Vicalloy II
Temperature = 4.2°K
PERCENT TRANSMISSION
102
100
98
96
94
92
-10
-8
-6
-4
-2
0
2
4
6
8
10
FWHM 1.852 mm / sec
ISOMER SHIFT -.0908 mm / sec
SOURCE-ABSORBER VELOCITY (mm / sec)

Figure 5

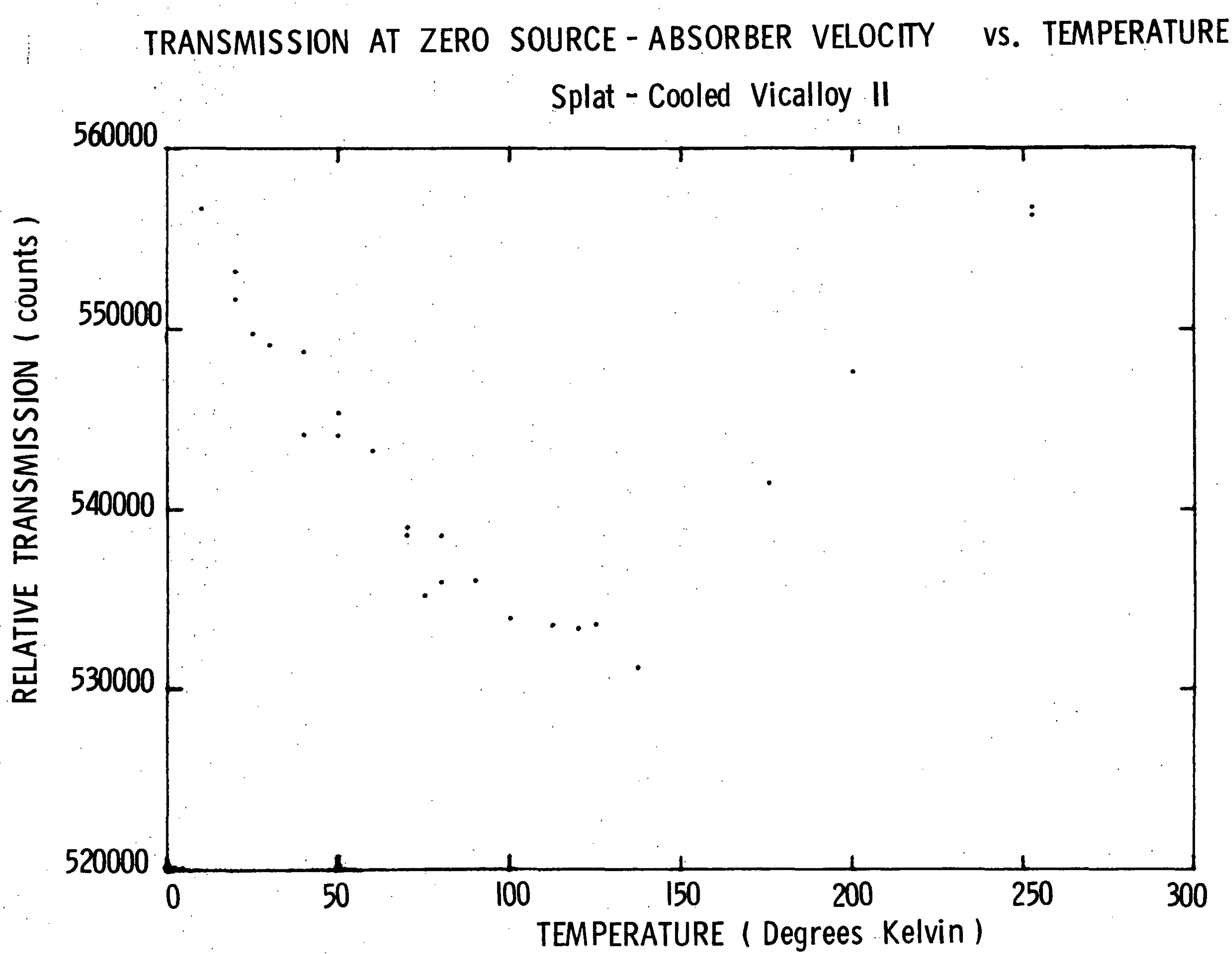

TRANSMISSION AT ZERO SOURCE - ABSORBER VELOCITY vs. TEMPERATURE
Splat - Cooled Vicalloy II
RELATIVE TRANSMISSION (counts)
560000
550000
540000
530000
520000
0
50
100
150
200
250
300
TEMPERATURE (Degrees Kelvin)

Figure 6

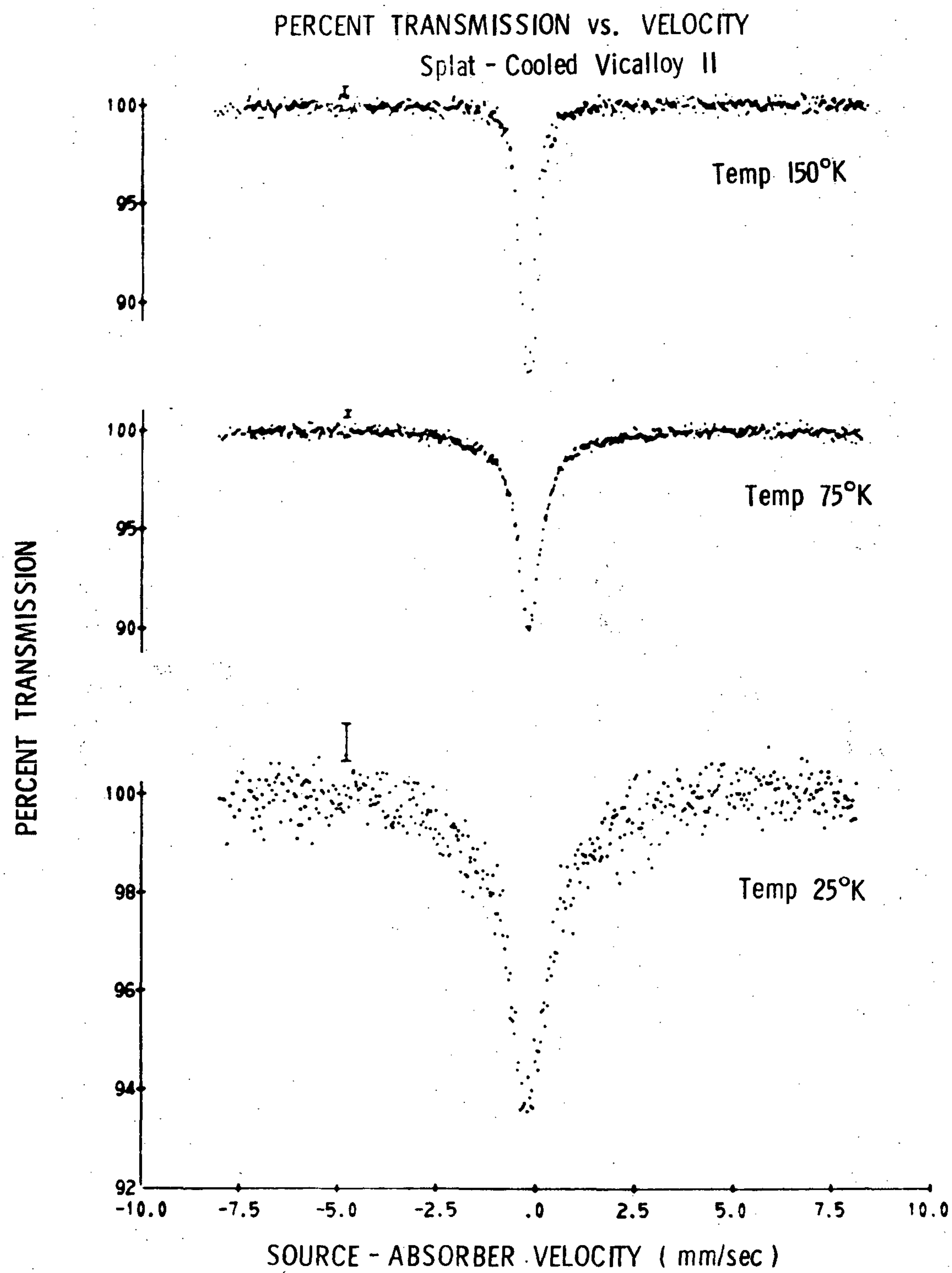

PERCENT TRANSMISSION vs. VELOCITY
Splat - Cooled Vicalloy II
Temp 150°K
Temp 75°K
Temp 25°K
PERCENT TRANSMISSION
100
95
90
100
95
90
100
98
96
94
92
-10.0
-7.5
-5.0
-2.5
.0
2.5
5.0
7.5
10.0
SOURCE - ABSORBER VELOCITY (mm/sec)

Figure 7